PB89-222632

Sources of Mutagenic Activity in Urban Fine Particles

(U.S.) Environmental Protection Agency
Research Triangle Park, NC

1989

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA/600/D-89/034		PB89-222632

4. TITLE AND SUBTITLE	5. REPORT DATE
SOURCES OF MUTAGENIC ACTIVITY IN URBAN FINE PARTICLES	6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
Robert K. Stevens, C.W. Lewis, T.G. Dzubay, L.T. Cupitt and J. Lewtas	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
U.S. Environmental Protection Agency Atmospheric Research and Exposure Assessment Laboratory Research Triangle Park, NC 27711	A101/C/83 11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Atmospheric Research and Exposure Assessment Laboratory Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC 27711	Symposium Paper 14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

16. ABSTRACT

Samples were collected during the winter of 1985-1986 in the cities of Albuquerque, NM and Raleigh, NC as part of a US Environmental Protection Agency study to evaluate methods to determine the emission sources contributing to the mutagenic properties of extractable organic matter (EOM) present in fine particles. Data derived from the analysis of the composition of these fine particles served as input to a multi-linear regression (MLR) model used to calculate the relative contribution of wood burning and motor vehicle sources to mutagenic activity observed in the extractable organic matter. At both sites the mutagenic potency of EOM was found to be greater (3-5 times) for mobile sources when compared to wood smoke extractable organics. Carbon-14 measurements which give a direct determination of the amount of EOM that originated from wood burning were in close agreement with the source apportionment derived from the MLR model.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSFIED	25
	20. SECURITY CLASS *(This page)* UNCLASSFIED	22. PRICE A03

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

REPRODUCED BY
U.S. DEPARTMENT OF COMMERCE
NATIONAL TECHNICAL INFORMATION SERVICE
SPRINGFIELD, VA. 22161

NOTICE

This document has been reviewed in accordance with U.S. Environmental Protection Agency policy and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

EPA/600/D-89/034

SOURCES OF MUTAGENIC ACTIVITY IN URBAN FINE PARTICLES

Robert K. Stevens, C.W. Lewis, T.G. Dzubay, L.T. Cupitt
and Joellen Lewtas
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

ABSTRACT

Samples were collected during the winter of 1985-1986 in the cities of Albuquerque, NM and Raleigh NC as part of a US Environmental Protection Agency study to evaluate methods to determine the emission sources contributing to the mutagenic properties of extractable organic matter (EOM) present in fine particles. Data derived from the analysis of the composition of these fine particles served as input to a multi-linear regression (MLR) model used to calculate the relative contribution of wood burning and motor vehicle sources to mutagenic activity observed in the extractable organic matter. At both sites the mutagenic potency of EOM was found to be greater (3-5 times) for mobile sources when compared to wood smoke extractable organics. Carbon-14 measurements which give a direct determination of the amount of EOM that originated from wood burning were in close agreement with the source apportionment results derived from the MLR model.

INTRODUCTION

The U.S. EPA Integrated Air Cancer Project (IACP) is a long range research program whose objectives include developing procedures to assess the environmental risk of human exposure to cancer producing emissions from a variety of sources. Unlike past efforts to identify airborne carcinogens through the use of emission inventories, the IACP takes the approach of measuring the mutagenicity (a surrogate for carcinogenicity) directly, and apportioning this property to source types.

The mutagenic activity of extractable organic matter (EOM) (EOM is defined as the non-volatile organic material present in fine particles that is extracted in dichloromethane, using a Soxhlet extraction procedure) has been used as a screening tool (Lewtas, 1988) to indicate the potential carcinogenicity of a number of primary emissions from selected combustion sources. There has been a limited attempt to estimate the potential mutagenic impact of combustion emissions on humans combining conventional predictive transport models with mutagenicity data on source emissions (Lewtas et al., (Review). The absence of both reliable emission inventories and knowledge on mechanisms of transport and transformation of chemical species responsible for mutagenic activity suggest that a better approach would be to apply receptor modeling (Lewis et al., 1988). Receptor models are not constrained by these limitations since they have the potential to use the chemical properties of aerosols measured at the source and receptor to calculate the source's contribution to mass or bioactivity of aerosols collected at the receptor.

To test the receptor modeling approach field studies were simultaneously conducted at two geographic locations, Albuquerque, NM and Raleigh, NC, during the 1984-1985 winter heating season. Residential wood burning and mobile source emissions were the only known significant contributors to the fine particle (EOM) content in both airsheds. The data collected from these two field locations were then used to calculate and validate the relative

contribution of mutagenic activity in ambient aerosols from the two sources. The receptor modeling results obtained from the air quality data collected at these two locations are discussed in this paper.

EXPERIMENTAL

Site Descriptions. In 1985 the EPA collected air quality data and samples in two urban locations in Albuquerque and Raleigh. Two primary sampling sites were established at each city. In each city, one of the two sites was located in a residential neighborhood impacted by wood smoke emissions. Numerous single family dwellings with operating fireplaces or woodstoves surrounded these primary sampling sites. Detailed description of the ambient monitoring sites is reported elsewhere (Highsmith et al., 1987). The data reported herein is based on the samples collected at the Albuquerque and Raleigh primary woodsmoke sampling sites.

Sampling Schedule. Aerosol samples were collected in Albuquerque and Raleigh for 12 hour periods beginning 7 A.M. and 7 P.M. in order to represent daytime and nighttime conditions. A complete description of the monitoring performed at these sites has been described elsewhere (Highsmith et al., 1987). In Albuquerque, sampling began January 12th and was nearly continuous until February 20th. In Raleigh, samples were collected intermittently from January 15th until March 20th, 1985. Most

samples in Raleigh were collected when the meteorological forecasts predicted cold periods. Since residential wood burning would be most prevalent during colder periods, higher ambient concentrations of EOM originating from wood stoves would be anticipated in fine particle samples collected during these periods.

Sampling and Analysis. Table 1 is a list of sampling systems and the types of analysis (Courtney et al., 1982; Stevens, 1985; Stevens et al., 1978; Klouda et al., 1987; Williams et al., in press; Ames et al., 1975; Claxton et al., 1987; Bernstein et al., 1982) performed on samples collected in Albuquerque and Raleigh in support of the receptor modeling phase of this study.

After sample collection for ^{14}C analysis the quartz filters were shipped in containers, kept at ~5°C, to the EPA Laboratories at Research Triangle Park for storage, analysis, and documentation. The Teflon coated glass filters were used to obtain the dichloromethane extracts for measuring mutagenic activity and the mass of EOM. A solvent and extraction methods study describing the selection of an optimum solvent, (dichloromethane) and extraction technique is reported elsewhere (Williams et al., in press). These filters were stored at -70°C after collection for approximately 6 months prior to extraction to preserve the sample integrity.

Table 1. Aerosol Sampling Equipment, Collection Conditions and Analyses Performed In Support of Receptor Modeling

Location	Samplers		Flow Rate,	Per-iods*	Filter	Measurements	
	Type	No.	L/min		Media	Species	Method
Albuquerque							
	Automated Sierra (Modified)	1	23.0	72	Teflon Quartz	mass Carbon	gravimetric pyrolysis
	Beckman Dichotomous	1	16.7	72	Teflon	mass elements	gravimetric by XRF
	PM-10 Hi-vol	2	1000	72	Quartz	Carbon C-14	pyrolysis Accelerator
	Hi-vol with 2.5 μm impactor	2	1000	72	Teflon coated glass	mutagens	Ames test
Raleigh							
	Sierra dichotomous sampler	1	16.7	57	Teflon	mass elements	gravimetric by XRF
	Hi-vol with Sierra Impactor Inlet (2.5 μm)	2	1000	57	Quartz	Carbon C-14	pyrolysis Accelerator
	Hi-vol with Anderson Impactor (2.5 μm)	2	1000	57	Teflon coated glass	mutagens	Ames test

*Duration of all periods is 12 h; 44 of the 72 sampling periods in Albuquerque, NM and 40 of the 57 periods in Raleigh, NC were used in the receptor modeling calculations.

The mutagenic activity of the methylene chloride extract was measured by first solvent exchanging with dimethyl sulfoxide, and assaying the organic matter in Salmonella typhimurium TA98 with

and without an Aroclor 1254 induced rat liver homogenate (+S9). The mutagenic activity was low in the absence of S9 and those results are not presented here. The assay was performed according to the standard Salmonella typhimurium plate incorporation assay (Ames et al., 1975) with minor modifications (Claxton, 1981 and Claxton et al., 1987). Data was analyzed and slope values were generated using the method of Bernstein et al. (1982). The uncertainty of these bioassay measurements, based on one-half the difference between duplicate determinations, averaged 14%.

Data from the analyzed samples were merged into a single data file for each site to be used in the receptor modeling calculations. ^{14}C Analysis. A sub-set of hi-volume fine particle samples collected on quartz filters were selected and sent to the National Bureau of Standards (NBS) for ^{14}C analysis. Details of the analysis procedure have been previously described (Klouda et al., 1987). Due to the relatively high cost of ^{14}C analysis only 13 of these samples were analyzed by NBS. The criterion for selecting these samples was based on having sufficient mass of EOM in each sample for ^{14}C analysis. These were selected from the 84 samples from Raleigh and Albuquerque that were used in the receptor model calculations described below. Results from the ^{14}C analysis were then compared to source apportionment results obtained statistically with a multiple linear regression (MLR) model. Additional ^{14}C measurements were made on composite samples and selected fractions from extracts of samples collected in this

study. Results of these measurements are reported elsewhere (Klouda et al., 1987).

RESULTS

Air Quality Data. Average concentrations of the key unique tracer species [Pb for mobile sources and K for wood burning (Stevens, 1985)] used in the receptor model calculations measured in Raleigh and Albuquerque are shown in Table 2.

Table 2. Average Concentration of Tracer Species in $\mu g/m^3$ Used in Receptor Model Calculations for Albuquerque, NM and Raleigh, NC Studies

	Albuquerque		Raleigh	
Species	Day	Night	Day	Night
Pb	0.270	0.294	0.082	0.117
K*	0.025	0.105	0.072	0.175

K* = K corrected for soil contribution

These elemental concentrations were obtained by x-ray fluorescence analysis (Stevens et al., 1978) of fine particle aerosol collected on Teflon filters using a dichotomous sampler at each site. These data indicate that the K concentrations were higher at night than during the day. However, the average Pb concentrations for the paired day-night sampling periods at both sites were similar. Nighttime meteorological inversion conditions reduced the mixing

height of the atmosphere which resulted in concentrating the pollutant emitted below the inversion. This probably accounts for the similarities between the day-night Pb concentrations. This concentration pattern suggests mobile sources contributed about the same amount of fine particles during both the day and night while residential wood burning emissions were mainly at night. Receptor modeling results discussed later, tend to support this conclusion. Previous wintertime air quality data and receptor modeling results from studies performed in Albuquerque and Denver (Lewis and Einfeld, 1985; Lewis et al., 1986) are similar in many respects to the data and results reported here.

Receptor Modeling. The source apportionment of EOM was performed with a single element tracer MLR model of the form

$$EOM_i = a\ (\text{Woodsmoke tracer})_i + b\ (\text{Motor vehicle tracer})_i + c \quad (1)$$

where the subscripted quantities are measured simultaneously during each sampling period i, and the initially unknown coefficients a, b, and c, are determined by MLR using data from all the sampling periods together. The measured concentration of a tracer for sampling period i multiplied by its coefficient is the best estimate of the corresponding source's contribution to total EOM concentration during that sampling period. The intercept can be regarded either as the average contribution of additional unknown sources, or as a measure of inadequacy of a two-source model. For either interpretation c should be small (relative to the average value of EOM) for an acceptable model.

In previous work in Denver (Lewis and Einfeld, 1985) a soil-corrected fine particle potassium tracer was used for wood smoke:

$$K^*_i = K_i - Fe_i\ (K/Fe)_{soil} \qquad (2)$$

where the soil ratio was determined from the average K/Fe ratio in the coarse dichotomous samples. The soil ratio (K/Fe) in Denver was found to be 0.68 while in Albuquerque and Raleigh the ratios were 0.45 and 0.42 respectively. For our MLR model we required a minimum of 30-50 observations to minimize the uncertainties in the source apportionment results.

The MLR equation set the dependent variable as either the mass of EOM_i in $\mu g\ m^{-3}$ or mutagenic activity expressed as the number of revertants per cubic meter (R_i) measured for each sampling period i. The independent variables were K^* and Pb, and their concentrations are expressed in $\mu g\ m^{-3}$. The utility of this approach has been demonstrated, previously, with the Albuquerque data set (Lewis et al., 1988) and data collected in Denver, CO (Lewis and Einfeld, 1985). For the Albuquerque data set, which consisted of 18 daytime and 26 nighttime observations, the linear regression results for EOM, [equation (1)] and mutagenic activity (R_i) [equation (2)] were as follows:

(Observations = 18 day and 26 night)

$$EOM = (204 \pm 8)\ K^*_i + (10.6 \pm 3)\ Pb_i + (1.0 \pm .9) \qquad (3)$$

$$r = 0.98$$

$$R_i = (258 \pm 27)\ K^*_i + (39 \pm 10)\ Pb_i + (3 \pm 3) \qquad (4)$$

$r = 0.90$

The standard error for the K-coefficients and intercepts are relatively small while the uncertainty in the Pb coefficients are nearly 30%. The larger uncertainty in the Pb coefficient is, in part, associated with the larger number of night time samples used in the MLR. However, the large multiple correlation coefficient (r) is an indication of the correctness of the model. Additional details related to the MLR model have been discussed previously (Lewis et al., 1988). Figure 1 is a graphical representation of the average contributions of the sources to the ambient concentrations of EOM and mutagenicity measured at the Albuquerque site, derived from equations (3) an (4).

For the Raleigh data both the high Br-Pb correlation coefficient (0.98) and Br/Pb ratio (.27) were similar to those found in Albuquerque, which strongly supports the assumed motor vehicle origin of these elements. However, when we attempted to perform our linear regression analysis on the Raleigh data set the coefficient for Pb had a large uncertainty. The samples collected in Raleigh and used in the MLR were dominated by wood smoke because the selection of samples for mutagenic analysis used in the receptor model calculations was based on the sample having sufficient mass of EOM available for mutagenic analysis. At the Raleigh site, this meant that the selected samples for MLR calculations were dominated by residential wood burning activities since EOM concentrations were elevated only during cold weather

events, a period when wood burning is most prevalent. Therefore, greater than 90% of the EOM for this selected data set was due to wood smoke emissions, making derivation of a reliable Pb coefficient difficult. Because of this problem we decided to use the coefficient for Pb derived from our Albuquerque study to obtain acceptable MLR results for the Raleigh data set. Since the two studies were contemporary we felt justified in using the Albuquerque derived Pb coefficient. Below are the linear regression results for Raleigh:

(observations = 14 day and 26 night)

$$EOM_i = (112 \pm 4)\ K^*_i + (10.6 \pm 3)\ Pb_i + (0.1 \pm .8) \quad (5)$$

$$r = 0.98$$

$$R_i = (88 \pm 7)\ K^*_i + (39 \pm 10)\ Pb + (1 \pm 1) \quad (6)$$

Again we note the relatively small uncertainty in our K^* coefficient, small intercepts and high correlation coefficients (r) for EOM and bioactivity (R_i). These results suggest our use of the Albuquerque, Pb coefficient in the MLR analysis of the Raleigh data did not substantially degrade our source apportionment.

The MLR coefficient for K^* derived from the Raleigh EOM data was significantly smaller than the coefficient derived in Albuquerque. This difference in the K^* coefficient is consistent with the variations of K emitted by the different types of wood burned in these two cities. In Raleigh, oak or other hardwoods are the major sources of fuel for wood stoves. Potassium

emissions from hardwoods are significantly greater than from softwoods (pine) (Stevens, 1985). Pinon pine is the principal wood used in Albuquerque; therefore, lower average ambient K concentrations would be expected and were found in Albuquerque than in Raleigh.

A jackknife procedure (Lewis et al., 1986) was also applied to these data sets to obtain non-parametric estimates of the regression coefficients and their errors. In this procedure, the set of regression results obtained from separately deleting each case is combined to give new estimates of the regression coefficients and their standard errors that are nonparametric, i.e., not dependent on the normality assumptions that underlie classical regression. For the 44 observations in Albuqerque and the 40 observations in Raleigh the jackknife estimates were virtually the same as the values obtained from the standard MLR analysis. Figure 2, giving average source contributions for the Raleigh site, is analogous to Figure 1 for Albuquerque.

Mutagenic Contribution and Potency of Sources. The ambient measurement-derived potencies of wood smoke and motor vehicle emissions for Albuquerque and Raleigh are derived from the linear regression equations (3-6). They are obtained from the ratio of the mutagenic activity to EOM for each source, and results are summarized in Table 3 and graphically represented Figures 1 and 2.

Table 3. Average Ambient Concentrations and TA98 +S9 Mutagenic Potencies Attributed to Woodsmoke and Mobile Sources.

	Wood smoke	Mobile sources	Other sources	Total
Albuquerque Results				
EOM, μg/m³	15 ± 1	3 ± .9	1 ± 0.9	19
Mutagenicity, rev/m³	19 ± 2	11 ± 3	3 ± 3.1	32
Potency, rev/μg EOM	1.3 ± .3	3.7 ± 1.5		
Raleigh Results*				
EOM, μg/m³	16 ± .5	1. ± .3	0.1 ± 1	17
Mutagenicity, rev/m³	12 ± .9	4. ± 1	1 ± 1	18
Potency, rev/μg EOM	.78 ± .07	3.7 ± 1.5		

*Pb Coefficients were from Albuquerque results

These results show the contribution of "other" sources to total mutagenicity to be small, indicating that mutagenic activity observed for samples from Albuquerque and Raleigh is almost entirely due to emissions from mobile and residential wood burning sources. In Albuquerque mobile sources produced 3.7 revertants/μg EOM compared to 1.3 revertant/μg from residential wood burning. In Raleigh the wood smoke potency was found to be 0.8 revertants/μg EOM, comparable to the Albuquerque result. The motor vehicle potencies are identical, of course, because of the use of Albuquerque Pb coefficient in Raleigh. The potency values are consistent with the average of mutagenic tests of primary emissions from wood stoves and mobile sources and the types of wood fuels burned in these two cities (Lewtas, 1988).

Validation by ^{14}C. The ^{14}C content of ten fine particle samples

collected on quartz filters in Albuquerque and Raleigh were analyzed by a new micro analysis procedure developed by NBS (Klouda, 1987). The ^{14}C results provide a direct (non-statistical) means of measuring the fraction of aerosol carbon that originates from the new or vegetative carbon (Currie et al., 1981; Currie et al., 1983). Wood burning is assumed to be the source of vegetative carbon since there are no other known sources of fine particle ^{14}C, which is a reasonable assumption for samples collected in the winter in Raleigh and Albuquerque. Table 4 shows a comparison of ^{14}C and MLR results.

The MLR wood stove contribution results for Albuquerque were derived using the total carbon measurements (C_i in $\mu g\ m^{-3}$) obtained by a combustion (Stevens, 1985) method along with Pb and K data. The MLR equation for C_i was

$$C_i = (190 \pm 9)\ K^*_i + (14.1 \pm 3.7)\ Pb_i + (1.4 \pm 1.1) \quad (7)$$

$r = 0.97$ for 44 observations; Pb and K* expressed in $\mu g\ m^{-3}$. The percentage woodsmoke values for MLR used to compare with the ^{14}C measurements is simply the ratios of $(190 \pm 9)\ K^*_i$ to C_i.

For samples collected in Raleigh the total carbon measurements were not reproducible due to problems associated with the combustion apparatus. However, since the carbon content of the sample was primarily extractable organics (>90%) we chose to use the linear regression coefficients obtained for EOM to calculate the % woodsmoke carbon. Correspondingly, the MLR percentages shown for Raleigh are obtained from the ratios of (112

± 4) K^*_i to EOM_i.

Table 4. Percent of Fine Particle Carbon from Wood Burning

Location and Sampling Period	^{14}C**** Results*	MLR Results
Albuquerque Results		
PM	75 ± 6	69 ± 10
PM	66 ± 3	75 ± 11
AM	58 ± 6	52 ± 12
PM	80 ± 5	98 ± 11
AM	61 ± 12	49 ± 13
PM	66 ± 3	61 ± 10
Raleigh Results**		
PM***	84 ± 5	94 ± 9
PM***	89 ± 5	94 ± 9
PM	98 ± 5	94 ± 10
PM	73 ± 7	88 ± 9

PM = 7 PM-7AM; AM=7AM-7PM
*Total Carbon Results
**Mass of extractable organics used in MLR calculations for Raleigh, NC samples
***Duplicate ^{14}C analysis
****The ^{14}C results have been corrected for the age of the wood.

The ^{14}C percentages are the NBS-measured fractions of "contemporary" carbon in each sample, divided by a correction factor of 1.10, which relates to the age of the wood (Currie et al., 1981), estimated to be about 60 years in both airsheds. For wood harvested in 1985 the maximum possible correction is 1.15, which would occur for 31 year old wood (Currie, private communication). In virtually every case the ^{14}C and MLR results overlap within the uncertainty limits.

Another means of evaluating the reasonableness of the MLR source apportionment results is to compare the % Wood Smoke Carbon derived from ^{14}C measurements with soil corrected ^{+}K concurrently obtained in Albuquerque and Raleigh. The results of this comparison is graphically represented in Figure 3. From the slopes of two data sets plotted in Figure 3, we determine directly the coefficients which when multiplied by $^{*}K$ concentration gives the contribution of fine particle carbon derived from wood burning. These values were 200 ± 11 and 110 ± 8 for Albuquerque and Raleigh respectively and are within ± 5% of the values obtained from the MLR model calculations.

DISCUSSION

A feature of this study that deserves discussion is the determination and source apportionment of the mutagenic potency of the EOM samples from Raleigh and Albuquerque. The samples collected in this study represent the composite contributions of literally thousands of homes and mobile sources, yet the mutagenic potency of the two major source types can be deduced. The mutagenic potency attributed to the mobile sources emissions in Albuquerque shown in Table 3 was found to be 3.7 revertants/μg. In addition, the mobile source emissions mutagenic potency obtained in this study was 3 times larger than the value calculated for the wood burning emissions. It is important to note these values shown in Table 3 are consistent with mutagenic tests of primary emissions from wood stoves and mobile sources

performed by a number of investigators (Claxton, 1983; Levtas, 1988). Thus from a comparison of these ambient results with the source mutagenic tests one may conclude the net effect of atmospheric chemical transformations of the primary emissions is small.

Various researchers have examined the effects of atmospheric transformations upon the mutagenicity of the wood smoke particulate emissions (Kamens et al., 1984; Bell and Kamens, 1986). The effects have been varied, sometimes increasing and sometimes decreasing the mutagenicity of the bulk sample. The agreement between the calculated mutagenic potencies in the air shed and the observed values for the direct source emissions suggests that transformations, if they did occur, had little net effect upon mutagenicity of the particulate involved. This result is consistent with smog chamber irradiation experiments (Kleindienst et al., 1986) also conducted as part of the IACP, which found that simulated atmospheric transformations had little or no effect upon the mutagenicity of aerosols.

A key feature of this receptor modeling approach was the fact that the study was performed without knowledge of the emission rates or populations, or mechanisms of transport or transformations of the fine particles. However, data on the K and Pb present in the primary emissions from residential wood burning and mobile sources respectively, are a necessary ingredient.

Finally the new technique for ^{14}C analysis by accelerator

mass spectrometry for 50-500 μg carbon (Verkouteren et al., 1987; Slota et al., 1987) was applied to samples collected in Raleigh and Albuquerque. These results serve as an independent direct check on MLR results and provides a means of validating our statistical model.

Disclaimer. The information in this document has been funded by the United States Environmental Protection Agency. It has been subject to Agency review and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

ACKNOWLEDGEMENTS

The authors express appreciation for the contributions made to the field sampling, chemical and biochemical analysis and data interpretation provided by the following colleagues in support of this Integrated Air Cancer Project: Ross Highsmith, Roy Zweidinger, Lloyd Currie, Larry Claxton, and Ralph Baumgardner. Without their contributions preparation of this report would not have been possible.

REFERENCES

AMES, B.N., McCANN, J. and YAMASAKI, E. (1975). Methods for detecting carcinogens and mutagens with the Salmonella/mammalian microsome mutagenicity test. Mutation Res. 31347-364.

BELL, D.A. and KAMENS, R.M. (1986). Photodegradation of wood smoke mutagens under low NO_x conditions. Atmos. Environ. 20: 317.

BERNSTEIN, L., KALDOR, J., McCANN, J. and PIKE, M.C. (1982). An empirical approach to the statistical analysis of mutagenesis

data from the Salmonella test. Mutation Res. 97:267-281.

CLAXTON, L.D. (1981). Mutagenic and Carcinogenic potency of diesel and related environmental emissions: Salmonella bioassay. Environment International 5:389-391.

CLAXTON, L.D. (1983). Characterization of automotive emissions by bacterial mutagenic bioassay: A review. Environ. Mutagen. 5:609-631.

CLAXTON, L.D., ALLEN, J., AULETTA, A., MORTELMANI, K., NESTMAN, E. and ZEIGLER, E. (1987). Guide for the Salmonella typhimurium/mammalian microssome tests for bacterial mutagenicity. Mutation Res. 189:83-91.

COURTNEY, W.J., SHAW, R.W. and DZUBAY, T.G. (1982). Precision and accuracy of a β gauge for aerosol mass determination. Environ. Sci. Technol. 16:236.

CURRIE, L.A. Private communication.

CURRIE, L.A., KLOUDA, G.A., CONTINETTI, R.E., KAPLAN, I.R., WONG, W.W., DZUBAY, T.G. and STEVENS, R.K. (1983). On the origin of carbonaceous particles in American cities: Results of radiocarbon "dating" and chemical characterization. Radiocarbon 25:603.

CURRIE, L.A., KLOUDA,G.A. AND GERLACH, R.W. (1981). Radiocarbon: Nature's tracer for carbonaceous pollutants. In: Residential Solid Fuels. Environmental Impacts and Solutions (J.A. Cooper and D. Malek, eds), pp. 365-385. Oregon Graduate Center Press, Beaverton, Oregon.

HIGHSMITH, R., BAUMGARDNER, R.E. HARRIS, B., LUMKIN, T., DRAGO, R., ZWEIDINGER, R. and MERRILL, R. (1987). The collection of wood combustion samples in Raleigh, NC and Albuqerque, NM, Proceedings EPA/APCA Symposium on Measurements of Toxic and Related Air Pollutants, Research Triangle Park, NC.

KAMENS, R.M., RIVES, T.D., PERRY, J.M., BELL, D.A., PAYLOR, R.F., GOODMAN, R.D. AND CLAXTON, L.D. (1984). Mutagenic changes in dilute wood smoke as it ages and reacts with ozone and nitrogen dioxide: An outdoor chamber study. Environ. Sci. Technol. 18:523.

KLEINDIENST, T.E., SHEPSON, P.B., EDNEY, E.O., CLAXTON, L.D. and CUPITT, L.T. (1986). Wood smoke: measurement of photooxidation products. Environ. Sci. Technol. 20:493.

KLOUDA, G., CURRIE, L., WISE, S., BENNER, B. and SHEFFIELD, A. (1987). Source apportionment of carbonaceous combustion products by micro radiocarbon measurements. Symposium on Measurement of Toxic and Related Air Pollutant, Research Triangle Park, NC.

LEWIS, C.W., BAUMGARDNER, R.E., STEVENS, R.K., CLAXTON, L.D. and LEWTAS, J. (1988). The contribution of woodsmoke and motor vehicle emissions to ambient aerosol mutagenicity. Environ. Sci.Technol. 22:968-971.

LEWIS, C.W., BAUMGARDNER, R.E., STEVENS, R.K. and RUSSWURM, G.M. (1986). Receptor study of Denver winter haze. Environ. Sci. Technol. 20:1126.

LEWIS, C.W. and EINFELD, W. (1985). Origins of carbonaceous aerosol in Denver and Albuquerque during the winter. Environ. Internat. 11:243.

LEWTAS, J. (1988). Combustion emissions: Characterization and comparison of their mutagenic and carcinogenic activity. In: Carcinogens and Mutagens in the Environment (H.F. Stich, ed), Vol. 5, pp. 59-74. CRC Press, Boca Raton, Florida.

LEWTAS, J., ALFHEIM, I. and LOFROTH, G. (In Review). Contribution of source emissions to the mutagenicity of ambient urban air particles.

SLOTA, JR., P.J., JULL, A.J.P., LINICK, T.W. and TOOLIN, L.T. (1987). Preparation of small samples for ^{14}C accelerator targets by catalytic reduction of CO. Radiocarbon 29:303.

STEVENS, R.K. (1985). Sampling and analysis for use in source apportionment studies to determine impact of wood burning on fine particle mass. Environ. Internat. 11:271.

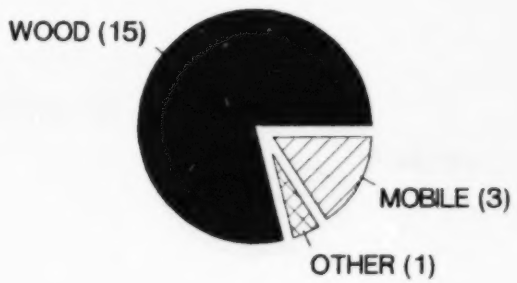
STEVENS, R.K., DZUBAY, T.G., RUSSWURM, G.M. and RICKEL, D. (1978). Sampling and analysis of atmospehric sulfates and related species. Atmos. Environ. 12(1-3):55.

VERKOUTEREN, R.M., KLOUDA, G.A., CURRIE, L.A., DONAHUE, D.J., JULL, A.J.P. and LINICK, T.W. (1987). Preparation of microgram samples on iron wool for radiocarbon analyzer via acceleration mass spectrometry; a close-system approach. Nucl. Instr. and Methods.

WILLIAMS, R., PASLY, T., WARREN, S., ZWEIDINGER, R., WATTS, R., STEAD, A. and CLAXTON, l. (In Press). Selection of a suitable extraction method for mutagenic activity from woodsmoke-impacted air particles. Int. J. of Environ. Anal. Chem.

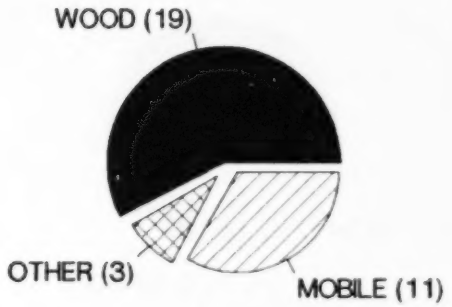
AVERAGE EXTRACTABLE ORGANICS CONCENTRATIONS

$\mu g / m^3$



AVERAGE MUTAGENIC CONCENTRATIONS

revertants / m^3



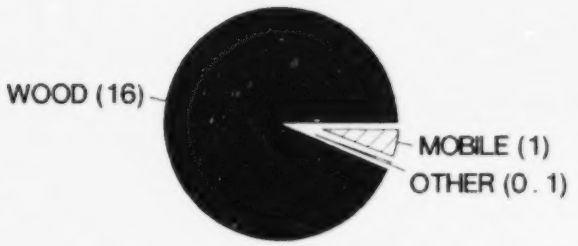
+S9

Figure 1. Graphical representation of the results of MLR model source apportionment of the average mass of extractable organic concentrations and average mutagenic activity derived from samples collected in Albuquerque, NM during the winter of 1984-1985.

Fig 1

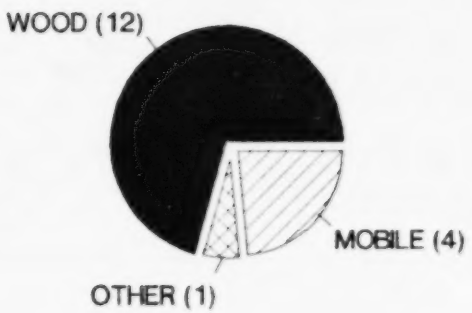
AVERAGE EXTRACTABLE ORGANICS CONCENTRATIONS

$\mu g / m^3$



AVERAGE MUTAGENIC CONCENTRATIONS

revertants / m^3



+S9

Figure 2. Graphic representation of the results of MLR model source apportionment of the average mass of extractable organic matter and average mutagenic activity derived from samples collected in Raleigh, NC during the winter of 1984-1985.

FIG 2

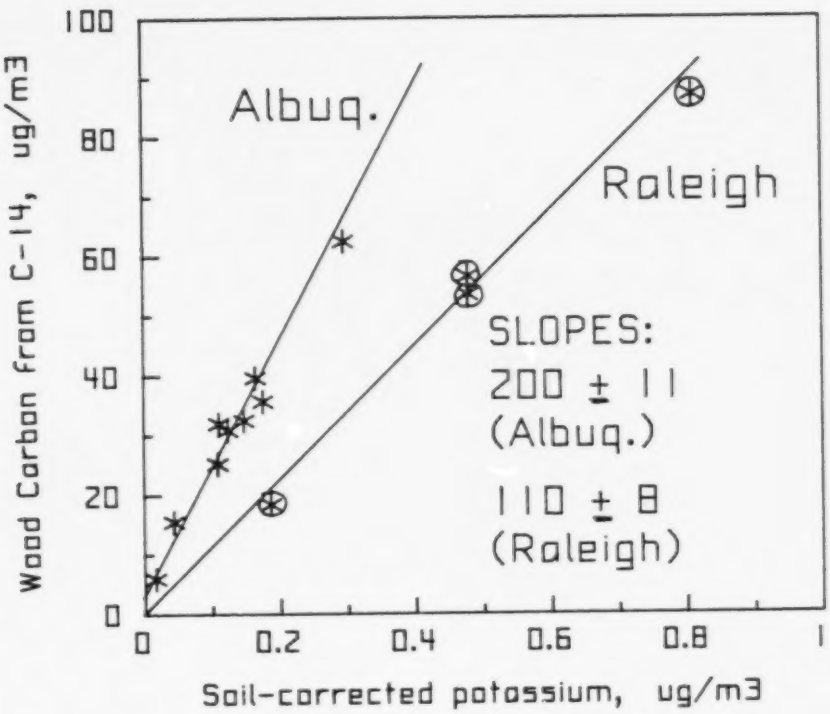


Figure 3. Soil corrected potassium in ug/m^3 plotted as a function ug/m^3 of wood carbon emissions from residential as determined from ^{14}C measurements from fine particle samples collected in Albuquerque, NM and Raleigh, NC.

Fig 3